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Thermodynamic features in the H-T plane of superconducting UBe_{13}

B. Ellman and T. F. Rosenbaum
The James Franck Institute and Department of Physics, The University of Chicago, Chicago, Illinois 60637

J. S. Kim and G. R. Stewart*
Department of Physics, The University of Florida, Gainesville, Florida 32611
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We present specific-heat C measurements of a high-quality polycrystal of UBe_{13} for $0.2 \leq T \leq 1.2$ K and $0 \leq H \leq 80$ kG. Field-sweep data, $C(H)$ at fixed T, reveal two broad peaks in the superconducting state for $T < 0.6$ K. The low-field feature occurs at $H \approx 20$ kG, independent of temperature. Conventional temperature scans, $C(T)$ at fixed H, presumably smear this feature beyond recognition.

The spin degrees of freedom play an essential role in heavy-fermion superconductivity. In UPt_3, not only may spin fluctuations provide the pairing mechanism, but the existence of antiferromagnetic order below T_c (with a magnetic moment $\sim 0.02\mu_B$)[1] allows the possibility of multiple superconducting phases.[2] Similarly, recent magnetic x-ray[3] and neutron-scattering[4] studies of URu_2Si_2 establish the microscopic coexistence of antiferromagnetism (with a magnetic moment $\sim 0.04\mu_B$) and superconductivity. Magnetic-susceptibility and heat-capacity measurements[5] on thoriated $CeCu_2Si_2$ show a significant bulk ordered moment coexisting with the superconductivity in that heavy-fermion system.

The situation for UBe_{13} is less settled. When doped with thorium, magnetic correlations arise[6] and for Th substitution between 2% and 4% two transitions are observed in the low-temperature specific heat.[7] For pure UBe_{13}, magnetostriction data[8] indicate the onset of antiferromagnetic order at $T_N = 8.8$ K $\sim 10T_c$, but there is no direct evidence to date for magnetic ordering below T_c. On the basis of muon spin resonance and lower critical-field measurements of superconducting UBe_{13}, Heffner *et al.*[9] rule out either magnetism or a second phase transition at low fields below T_c. In contrast, Rauchschwalbe[10] suggests two superconducting order parameters for UBe_{13} from an analysis of a break in the upper critical-field slope at $H \sim 20$ kG. We report here multiple thermodynamic signatures in pure UBe_{13} for $T/T_c < 0.6$ at $H \approx 20$ kG.

The key difference in our heat-capacity measurements is that we fix temperature T and sweep magnetic field H, in contrast to the conventional approach of fixing H and varying T. Varying the magnetic field introduces the technical complexity of recalibrating the thermometer attached to the sample at every one of typically 80 magnetic-field points. It provides, however, sensitivity to thermodynamic features when the phase boundary of interest is essentially parallel to the temperature axis in the H-T plane, features which would be smeared beyond recognition in a temperature sweep at fixed H. The specific heat $C(H)$ at fixed T has proved successful in revealing[11] sharp features in the H-T plane of UPt_3, even in a sample with one broad peak in C/T vs T at any H. Furthermore, the functional form of $C(H)$ for $T < T_c$ provides information about the nature of the excitation spectrum in the superconducting state.

The UBe_{13} sample used in our work is an arc-melted, high-purity polycrystal of mass 0.08 g. Preparation specifics have been provided elsewhere.[12] In zero magnetic field, the superconducting transition temperature $T_c = 0.925$ K, the Sommerfeld constant $\gamma = 1.0$ J/mol K, and the specific-heat jump at T_c, $\Delta C/C = 1.8$ (see Fig. 1). The experiments were performed using standard heat-pulse techniques in a helium dilution refrigerator for $0.2 \leq T \leq 1.2$ K and $0 \leq H \leq 80$ kG. As discussed above, the thermometer was a carbon chip whose magnetoresistance made it necessary to recalibrate it at every magnetic-field point during field sweeps. The heater was made of Au/Cr and was field independent to better than 0.03% over the entire magnetic-field range. The nuclear Zeeman contribution to the specific heat from Be (non-negligible for $H > 40$ kG at the lowest temperatures) has been subtracted from all the data.

We plot in Fig. 2 a series of specific-heat field sweeps for $T < T_c(H=0)$. At the higher temperatures it is possible to determine the upper critical field, delineated by the essentially field-independent response of $C(H)$ in the normal state. H_{c2} moves out of our magnetic-field window, however, by $T = 0.4$ K. At the lower temperatures ($T \leq 0.6$ K), $C(H)$ rises with increasing magnetic field as the vortices supply quasiparticles, dropping to the normal-state value of the specific heat with the approach

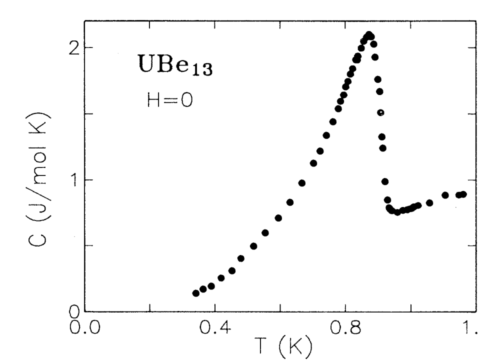

FIG. 1. Specific heat C in zero magnetic field at the superconducting transition.

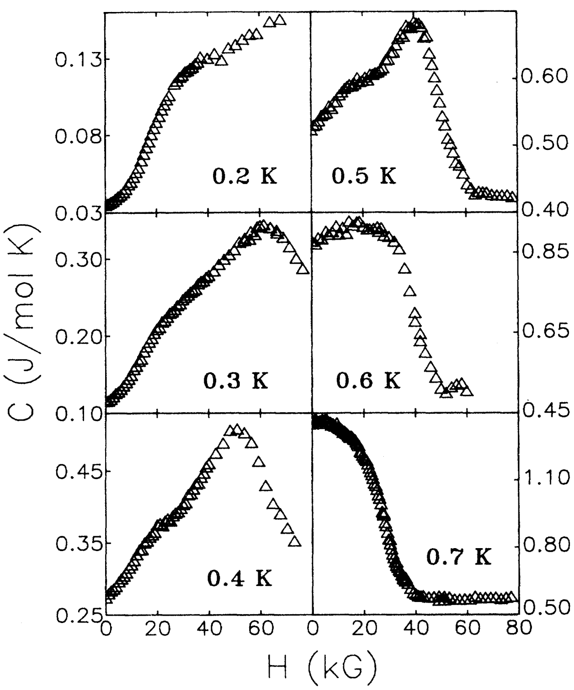

FIG. 2. Specific heat C of UBe_{13} as a function of magnetic field H at a series of temperatures $T < T_c(H=0)$. $C(H)$ is essentially independent of H in the normal state. Two peaks appear in the superconducting state for $T < 0.6$ K, most clearly distinguished at $T=0.5$ K.

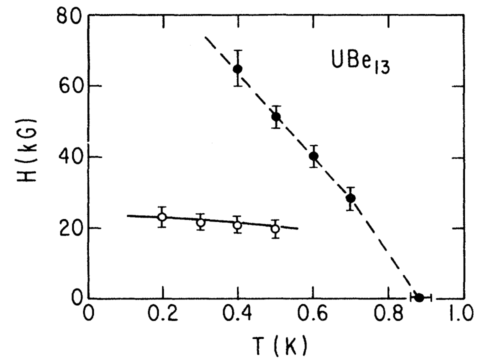

FIG. 3. Phase diagram in the magnetic-field temperature plane derived from Fig. 2. Solid circles correspond to values of H_{c2}. Open circles denote the position of the secondary maximum, as determined in a differential plot. Lines are guides to the eye. Lower boundary coincides with a change in slope of H_{c2} (Refs. 10, 13, and 14).

to H_{c2}. The quadratic form of $C(H)$ as $H \rightarrow 0$ apparent from $T=0.2$ and 0.3 K gives way to a more linear dependence on magnetic field for larger $T/T_c(H=0)$. At corresponding values of $T/T_c(H=0)$ in UPt_3 (Ref. 11) $C(H)$ is also proportional to H, suggesting a similar excitation spectrum.

The UBe_{13} data of Fig. 2 reveal an additional feature in $C(H)$ at an intermediate field, $H \approx 20$ kG, for $T < 0.6$ K. This secondary maximum can be observed most clearly at $T=0.5$ K, but it can be discerned as well at the three lower temperatures. We plot in Fig. 3 a putative phase diagram for UBe_{13} in the H-T plane based on the structure in the $C(H)$ curves. The solid circles correspond to the upper critical field, taken as the midpoint of the sharp rise in $C(H)$ at large H. The open circles mark the position of the partially buried peak at lower field as a function of T.

Although the origin of multiple thermodynamic features in magnetic field and temperature for UBe_{13} is not known, we can make some general observations regarding Fig. 3. First, it is clear from the nearly temperature-independent behavior of the lower boundary that cuts in $C(H,T)$ along H, as reported in this experiment, are required to see it. Second, the position of the lower boundary at $H \approx 20$ kG coincides with the change in slope of the upper critical-field curves reported previously by at least three groups.[10,13,14] Such a kink is consistent with our H_{c2} data of Fig. 3; additional points would be required to define it conclusively on the basis of specific-heat data. The boundary at $H \approx 20$ kG also coincides with an anomaly in the magnetostriction observed[8] at $T=0.6$ K. Third, the appearance of two features below $T=0.6$ K is at the same temperature posited for the emergence of a second superconducting order parameter in the Rauchschwalbe analysis[10] of H_{c2} data. However, that proposed phase diagram predicts a lower boundary at $H \approx 70$ kG, in contrast to the calorimetric features at $H \approx 20$ kG.

If there are indeed two distinct phases, then the question remains as to their nature, particularly in view of the precedents in other heavy-fermion compounds of the microscopic interplay of magnetism and superconductivity. The work of Heffner *et al.*[9] indicates that the lower phase cannot have a significant local magnetic moment ($\mu < 0.01\mu_B$). Many alternatives still exist, however, given the likely higher-order pairing in UBe_{13}. The possible role of quadrupolar coupling[15] may also predispose the system to a magnetic-field-induced crossover. The effects of symmetry-breaking fields other than the magnetic field, such as uniaxial stress,[16] may be able to help narrow the possibilities.

In summary, specific-heat measurements as a function of H at fixed T reveal both the transition into the normal state at $H_{c2}(T)$ and a secondary maximum at $H \approx 20$ kG for $T < 0.6$ K. The lower-field feature occurs at a kink in the slope of $H_{c2}(T)$ and suggests the possibility of distinct phases in superconducting UBe_{13}.

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*Also at The Universität Augsburg, Germany.
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